Development of measurement methods for nuclear materials (U/Pu) at enrichment and reprocessing facilities based on X-ray fluorescence, neutron coincidence counting and/or KX-ray absorption spectrometry

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Where?

ITU-Nuclear Safeguards and Security Unit

- **Analytical services**
- **Nuclear Safeguards R&D**
- **Laboratories in situ**

- **Radiometric techniques**
  - Gamma Spectrometry
  - K-edge Densitometry (KED)
  - X-ray fluorescence spectrometry (XRF)
  - COMbined Product Uranium Concentration and Enrichment Assay (COMPUCEA)
  - Neutron Coincidence Counting (NCC)
  - High Level Neutron Coincidence Counter (HLNC)
  - Alpha spectrometry
- **Chemical techniques**
  - Titration
  - Inducted couple plasma mass spectrometry (ICP-MS)
  - Thermal Ionization Mass spectrometry (TIMS)
  - Isotope dilution Thermal Ionization Mass spectrometry (IDMS)
  - Secondary ion mass spectroscopy (SIMS)

Methods

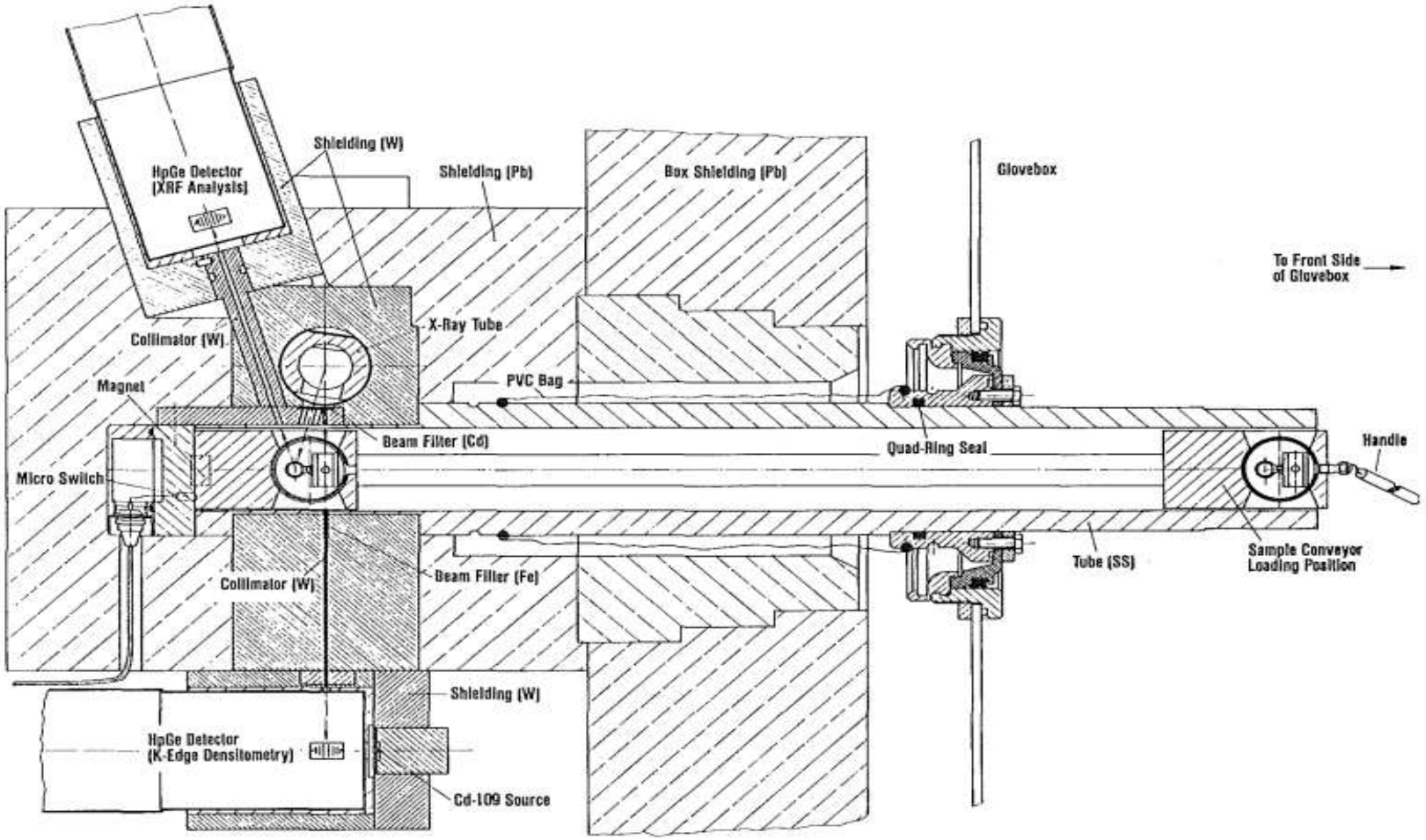
• At ITU the X-ray fluorescence (XRF) analysis is performed simultaneously with K-edge densitometry technique

•This hybrid method has been intensively operated at the ITU and in On-Site Laboratories (OSL) for safeguard purposes such as accountancy verification for many years.

•K-absorption edge spectrometry, (K-edge densitometry (KED)) is used to determine the uranium concentration.

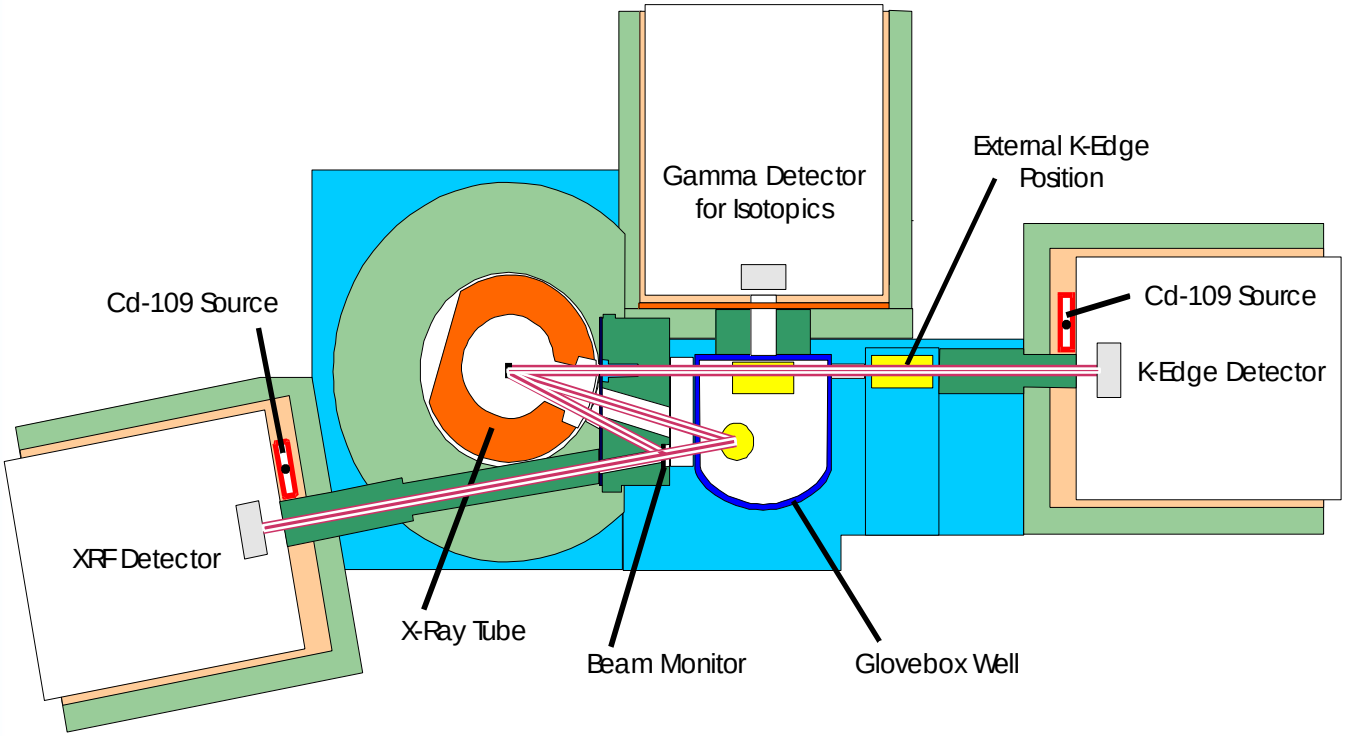
X-ray fluorescence (XRF) analysis is used to determine the U/Pu ratio (in concentrated U/Pu mixed solutions like the reprocessing input solutions)

X -ray beam geometry for KED and XRF in the Hybrid Instrument



H. Ottmar, H. Eberle *The Hybrid K-Edge/K-XRF Densitometer: Principles-Design-Performance*

Hybrid K-Edge/K-XRF densitometer (HKED)



In the XRF technique, relative detector efficiency is a key parameter in the analysis. Although it is relatively easy to determine it for U and Pu in a specific geometry, it might be more *problematic in presence of more exotic mixtures*. Furthermore, this relative efficiency is rather sensitive to the geometry and concentration of the investigated sample - except for small variations, where the effect might be neglected – and thus a new calibration would be required with adjustments on the analyzed sample.

A.N. Berlizov, D.A.Sharikov, H.Ottmar, H.Eberle, J. Galy, K.Luetzenkirchen, N. I. M. A 615 (2010) 127–135

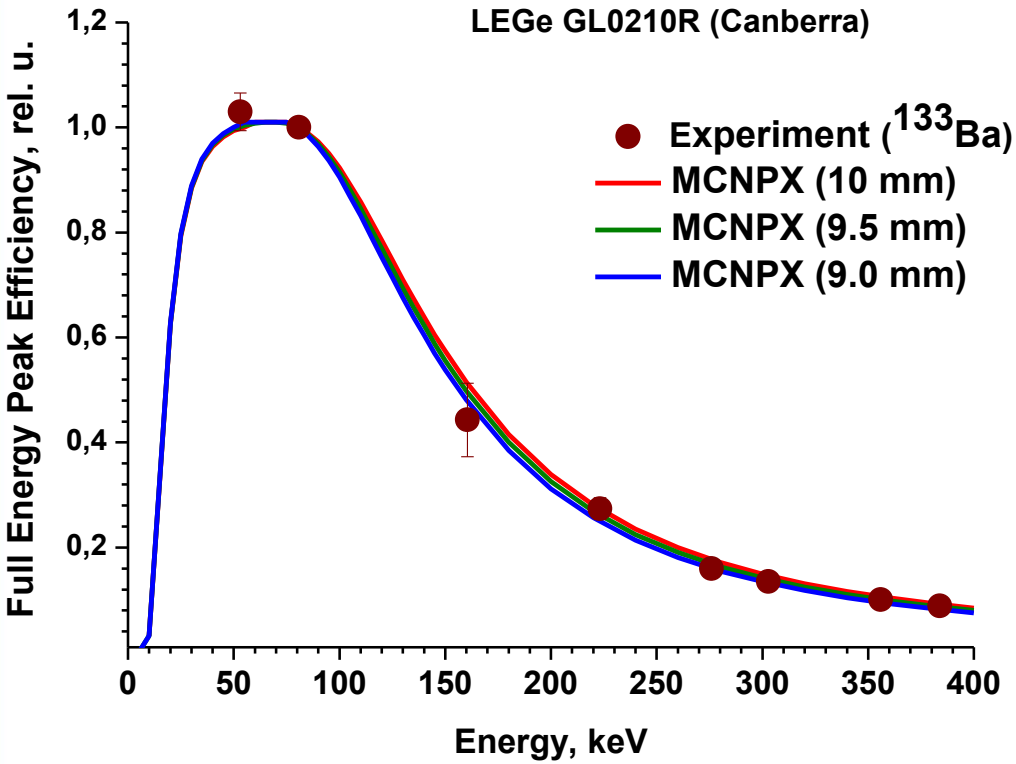
Monte Carlo Calculations

Goal: Develop models of hybrid K-Edge/K-XRF densitometers by means of MCNP (Monte Carlo N-Particle) Code so as to reliably calculate the response of HKED instruments in dependence of sample composition

Practical benefits:

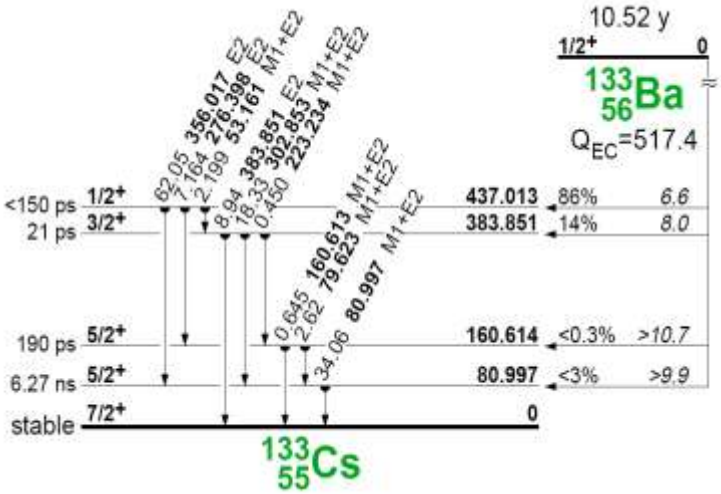
- ☺ Calibration efforts significantly reduced
- ☺ *The extension of the applicability of HKED method for unusual samples and non-standard sample matrices*
- ☺ Study special measurements effects representing potential sources of measurement biases

Detector intrinsic Full Energy Peak Efficiency calculation



Ba-133 certified reference source

- Ge cylinder - 500 mm2 × 10 mm
- Input window - 0.5 mm Al
- Photon pencil beam goes along the cylinder revolution axis



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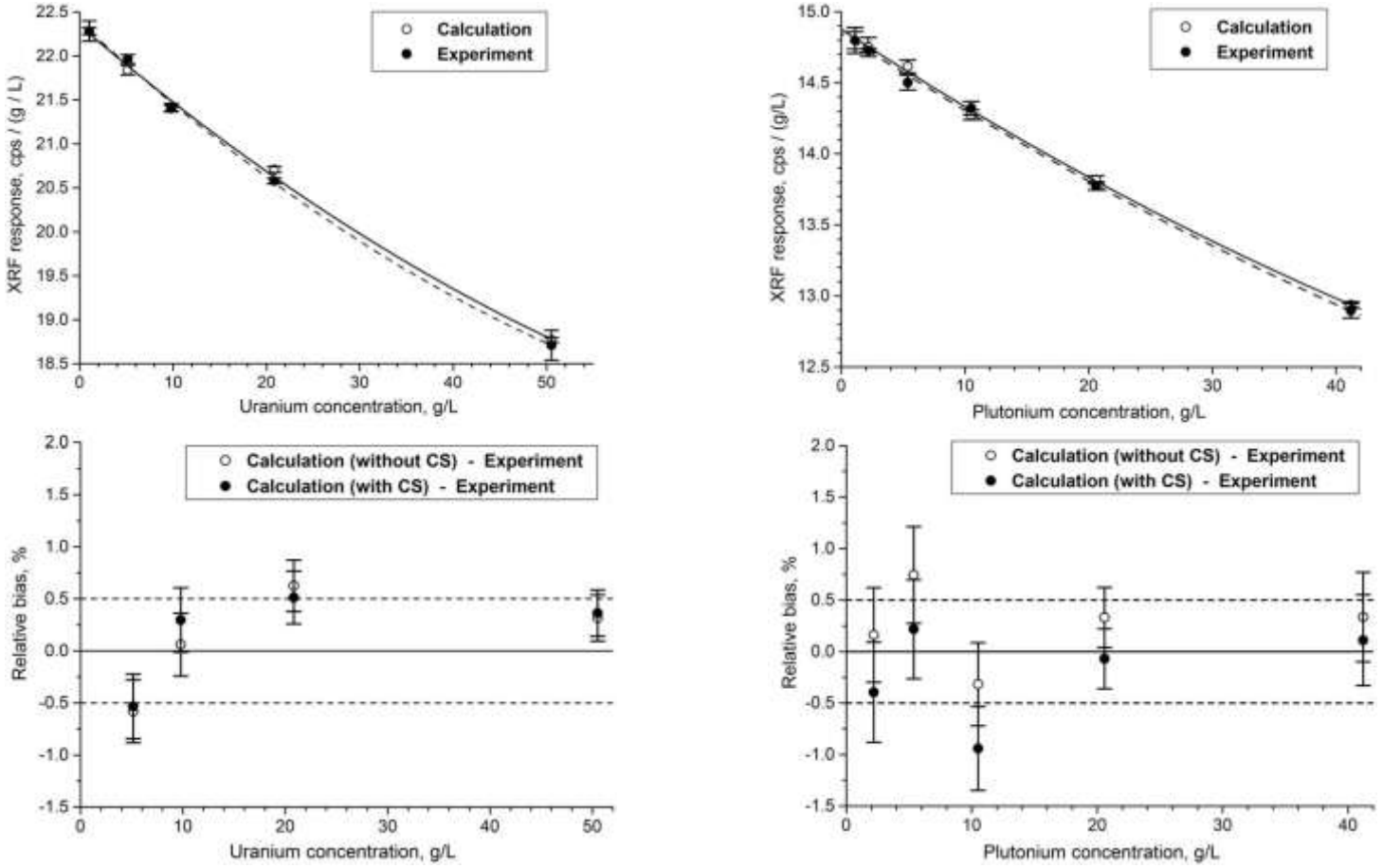
Monte Carlo Results



(a) (b)

XRF responses for the low U (a) and Pu (b) concentration measurements with a HKED-1 instrument. Top: experimental (dotted) and simulated (solid) XRF calibration curves. Bottom: percentage difference between the calculated and measured XRF calibrations.

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Summary

The Monte Carlo based mathematical simulation approach with respective MCNP models for the XRF branch of two different versions of HKED instruments currently in use was set up and validated. The achieved accuracy and speed of the simulations provide a firm basis for the further practical implementation of a consistent and time-efficient mathematical calibration approach for other applications like:

- the U/Pu element ratio measurement in the reprocessing input solutions and dissolved MOX product samples, where the ratio can be determined in principle solely based on the mathematical calibration of the instrument;

- the measurement of U and Pu concentrations in non-standard matrices (e.g. unusual molarity, presence of other actinides), where modelling is capable of providing accurate matrix corrections for the yields of characteristic KX-rays of actinides from the assayed solutions.

Thank you!